Verification Statement For Translation

I, KIM, Hyo-Jin, hereby declare that I am conversant in the Korean and the English languages and that I am the translator of the document attached and certify that to the best of my knowledge and belief the following is a true and correct English translation of the specification contained in Korean Patent Application No. 10-2002-0073961.

KIM, Hyo-Jin

Date:

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Application No.: 10-2002-0073961

Filing date: November 26, 2002

Applicant(s): SAMSUNG SDI CO., LTD.

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COMMISSIONER

(Translation)

APPLICATION FOR PATENT

5 RECEIPIENT: The Commissioner of the Korean Intellectual Property Office

Filing date: November 26, 2002

TITLE: POSITIVE ELECTRODE FOR LITHIUM-SULFUR BATTERY AND LITHIUM-SULFUR BATTERY

10 COMPRISING SAME

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GENERAL POWER OF ATTORNEY REGISTRATION NO.: 2001-041982-6

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The above identified application is duly submitted herewith pursuant to Article 42 of the Patent

30 Act.

ABSTRACT OF THE DISCLOSURE

[Abstract]

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The present invention relates to a positive electrode for a lithium-sulfur battery and a lithium-sulfur battery including the same, and the positive electrode includes a positive active material selected from elemental sulfur (S_8), a sulfur-based compound and mixtures thereof; a conductive material; a binder; and a V_2O_5 additive with a particle size D (v, 50%) of 1,000nm or less.

As described above, the positive electrode with the V_2O_5 additive with the critical particle size exhibits high ionic conductivity and decreased surface roughness, thereby increasing capacity from 1200mAh to 1289mAh, and improving cycle life from 60% to 90%.

[Key words]

V2O5, Particle size, Lithium sulfur battery, Positive electrode, Cycle life

[SPECIFICATION]

[TITLE OF THE INVENTION]

POSITIVE ELECTRODE FOR LITHIUM-SULFUR BATTERY AND LITHIUM-SULFUR BATTERY COMPRISING SAME

[DETAILED DESCRIPTION OF THE INVENTION]

[OBJECT OF THE INVENTION]

[FIELD OF THE INVENTION AND THE RELATED ART]

[Field of the Invention]

The present invention relates to a positive electrode for a lithium-sulfur battery and a lithium-sulfur battery comprising the same, and more particularly, to a positive electrode for a lithium-sulfur battery exhibiting good cycle life characteristics.

[Description of the Related Art]

The continued development of portable electronic devices has led to a corresponding increase in the demand for secondary batteries having both a lighter weight and a higher capacity. To satisfy these demands, the most promising approach is a lithium-sulfur battery with a positive electrode made of sulfur-based compounds.

Lithium-sulfur batteries use sulfur-based compounds with sulfur-sulfur bonds as a positive active material, and a lithium metal or a carbon-based compound which is one that can reversibly intercalate or deintercalate metal ions, such as lithium ions, as a negative active material. Upon discharging (i.e., electrochemical reduction), the sulfur-sulfur bonds are cleaved, resulting in a decrease in the oxidation number of sulfur (S), and upon recharging (i.e., electrochemical oxidation), the sulfur-sulfur bonds are re-formed, resulting in an increase in the oxidation number of the S, thereby generating the electrical energy and storing in the battery as chemical energy during charging, and is converted back to electrical energy during discharging.

With respect to specific density, the lithium-sulfur battery is the most attractive among the currently developing batteries since lithium has a specific capacity of 3,830 mAh/g, and sulfur has a specific capacity of 1,675 mAh/g. Further, the sulfur-based compounds are less costly than other materials, and they are environmentally friendly.

However, employing a positive electrode based on elemental sulfur in an alkali metal-sulfur battery system has been considered problematic. Although theoretically the reduction of sulfur to an alkali metal-sulfide confers a large specific energy, a very low percentage of utilization which indicates the amount of sulfur participated in the electrochemical redox reaction causes very low battery capacity.

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Thus, there have been various studies to improve the electrochemical redox reaction and to increase capacity.

[SUMMARY OF THE INVENTION]

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The present invention provides a positive electrode for a lithium-sulfur battery exhibiting good cycle life characteristic by controlling the roughness of the surface of the positive electrode using an additive with a critical particle size.

The present invention also provides a lithium-sulfur battery including the positive electrode.

[DETAILED DESCRIPTION OF THE INVENTION]

In one embodiment, the invention is directed to a positive electrode for a lithium-sulfur battery including a positive active material selected from elemental sulfur (S_8), a sulfur-based compound, or a mixture thereof; a conductive material; a binder; and a V_2O_5 additive with a particle size D (v, 50%) of 1,000nm or less.

The present invention also provides a lithium-sulfur battery including the positive electrode, a negative electrode including a negative active material selected from a material that is capable of reversibly intercalating or deintercalating lithium ions, a material that reacts with lithium ions to prepare a lithium-included compound, a lithium metal, and a lithium alloy, and an electrolyte

The present invention will be illustrated in more detail.

The present invention relates to a positive electrode for a lithium-sulfur battery using a V_2O_5 additive with a desired particle size, so that the positive electrode can provide batteries exhibiting good cycle life characteristics.

The positive electrode of the present invention includes the V_2O_5 additive with the particle size D (v, 50%) 1,000 nm or less, more preferably from 1 to 900 nm, even more preferably from 5 to 700 nm, and still more preferably from 10 to 500 nm. In this application, the term "particle size D (v, 50%)" means a particle size in which particles distributed with various size such as 0.1, 0.2, 0.3 3, 5, 7,10, 20, or 30 μ m are accumulated to 50 volume %. As the particle size D (v, 50%) decreases, ionic conductivity increases so that a small particle size is preferable. If the particle size D (v, 50%) is more than 1000nm, the surface roughness (Ra) of a produced positive electrode increases, making the surface of the positive electrode uneven, thereby deteriorating the capacity and particularly cycle life characteristics.

The particle size of the V_2O_5 additive may be controlled by a general mechanically mixing process such as a ball-milling process using a zirconia ball and a solvent for 3 to 24 hours. The solvent is any solvent that does not react with the metal oxide or metal sulfide, and useful solvents include isopropyl alcohol, ethyl alcohol, and methyl alcohol.

The V_2O_5 additive with the above particle size renders a decrease in the average surface roughness Ra of 5 μ m, and such a decreased surface roughness allows a decrease in interfacial resistance between a positive electrode and a separator, thereby decreasing internal resistance of batteries and providing good battery performance.

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U.S. Patent Nos. 6,238,821 and 6,210,831 disclose the use of a V_2O_5 additive in a positive electrode, but, these patents are silent on control of the particle size of V_2O_5 . and thus, it is well understood to one in the related art that the battery performance improvement effect of the present invention by decreasing the internal resistance cannot be realized by these patents. In addition, U.S. Patent No. 6,130,007 discloses a vanadium oxide positive active material with a particle size of 1000 nm or less, but this did not exhibit suitable capacity and cycle life.

The positive active material includes elemental sulfur (S_8), a sulfur-based compound, or a mixture thereof. The sulfur-based compound may be selected from Li_2S_n ($n \ge 1$), organic-sulfur compounds, and carbon-sulfur polymers ($(C_2S_x)_n$: x=2.5 to 50, $n \ge 2$).

The conductive material includes an electrically conductive material that facilitates the movement of electrons within the positive electrode. Examples of the conductive material include, but are not limited to, conductive materials such as graphite- and carbon-based materials, and conductive polymers. The graphite-based materials include KS 6 (manufactured by TIMCAL COMPANY), and the carbon-based materials include SUPER P (manufactured by MMM COMPANY), ketjen black, denka black, acetylene black, carbon black, and the like. Examples of the conductive polymer include, but are not limited to, polyaniline, polythiophene, polyacetylene, polypyrrole, and the like. The conductive material may be used alone or as a mixture of two or more of the above conductive materials.

A binder may be added to adhere the positive active material on a current collector. The binder may be poly(vinyl acetate), polyvinyl alcohol, polyethylene oxide, polyvinyl pyrrolidone, alkylated polyethylene oxide, crosslinked polyethylene oxide, polyvinyl ether, poly(methyl methacrylate), polyvinylidene fluoride, a copolymer of polyhexafluoropropylene and polyvinylidene fluoride (Trademark: KYNAR), polyethyl acrylate, polytetrafluoroethylene, polyvinyl chloride, polyacrylonitrile, polyvinyl pyridine, polystyrene, a derivative thereof, a blend thereof, or a copolymer thereof.

The positive electrode may further include a coating layer including a polymer, an inorganic material, or a mixture thereof.

The polymer may include polyvinylidene fluoride, a copolymer of polyvinylidene fluoride and hexafluoropropylene, poly(vinyl acetate), poly(vinyl butyral-co-vinyl alcohol-co-vinyl acetate), poly(methylmethacrylate-co-ethyl acrylate), polyacrylonitrile, polyvinyl chloride-co-vinyl acetate, polyvinyl alcohol, poly(1-vinylpyrrolidone-co-vinyl acetate), cellulose acetate, polyvinyl pyrrolidone, polyacrylate, polymethacrylate, polyolefin, polyurethane, polyvinyl ether, acrylonitrile-butadiene rubber, styrene-butadiene rubber, acrylonitrile-butadiene styrene, a sulfonated styrene/ethylene-butylene/styrene triblock copolymer, polyethylene oxide, or a mixture thereof.

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Examples of inorganic material include colloidal silica, amorphous silica, surface-treated silica, colloidal alumina, amorphous alumina, tin oxide, titanium oxide, titanium sulfide (TiS₂), vanadium oxide, zirconium oxide (ZrO₂), iron oxide, iron sulfide (FeS), iron titanate (FeTiO₃), barium titanate (BaTiO₃), and mixtures thereof. The organic material may be conductive carbon.

The positive electrode of the present invention is produced by the general procedure in which the positive active material, the conductive material, the binder, and the inventive a V_2O_5 additive with a particle size D (v, 50%) of 1,000nm or less are mixed in a solvent to prepare a composition (e. g. in the form of slurry), and the composition is coated on a current collector.

A lithium-sulfur battery of the present invention with the positive electrode includes a negative electrode and an electrolyte. The negative electrode of the lithium-sulfur battery includes a negative active material selected from materials in which lithium intercalation reversibly occurs, materials that react with lithium ions to form a lithium-containing compound, lithium metals, and lithium alloys.

The materials in which lithium intercalation reversibly occurs include carbon-based compounds. Any carbon-based compound may be used as long as it is capable of intercalating and deintercalating lithium ions. Examples of such carbon materials include crystalline carbon, amorphous carbon, and mixtures thereof.

Examples of the material that reacts with lithium ions to form a lithium-containing compound include, but are not limited to, tin oxide (SnO₂), titanium nitrate, and Si. The lithium alloys include alloys of lithium and a metal selected from Na, K, Rb, Cs, Fr, Be, Mg, Ca, Sr, Ba, Ra, Al, and Sn.

The negative electrode may include an inorganic protective layer, an organic protective layer, or a mixture thereof, on a surface of lithium metal. The inorganic protective layer includes Mg, Al, B, Sn, Pb, Cd, Si, In, Ga, lithium silicate, lithium borate, lithium phosphate, lithium phosphoronitride, lithium silicosulfide, lithium borosulfide, lithium aluminosulfide, or lithium phosphosulfide. The organic protective layer includes a conductive monomer, oligomer, or polymer selected from poly(p-phenylene), polyacetylene, poly(p-phenylene vinylene), polyaniline, polypyrrole, polythiophene, poly(2,5-ethylene vinylene), acetylene, poly(perinaphthalene), polyacene, and poly(naphthalene-2,6-di-yl).

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In addition, during charging and discharging of the lithium-sulfur battery, the positive active material (active sulfur) converts to an inactive material (inactive sulfur), which can be attached to the surface of the negative electrode. The term "inactive sulfur", as used herein, refers to sulfur that has no activity upon repeated electrochemical and chemical reactions so it cannot participate in an electrochemical reaction of the positive electrode, and the inactive sulfur on the surface of the negative electrode acts as a protective layer of the lithium negative electrode. Accordingly, inactive sulfur, for example lithium sulfide, on the surface of the negative electrode can be used in the negative electrode.

The electrolyte includes an electrolytic salt and an organic solvent.

The organic solvent may be a sole solvent or a mixed organic solvent with at least two components. The mixed organic solvent includes at least two groups selected from weak polar solvent groups, strong polar solvent groups, and lithium protection groups.

The term "weak polar solvent", as used herein, is defined as a solvent that is capable of dissolving elemental sulfur and has a dielectric coefficient of less than 15, and the weak polar solvent is selected from aryl compounds, bicyclic ether, and acyclic carbonate compounds. The term "strong polar solvent", as used herein, is defined as a solvent that is capable of dissolving lithium polysulfide and has a dielectric coefficient of more than 15, and the strong polar solvent is selected from bicyclic carbonate compounds, sulfoxide compounds, lactone compounds, ketone compounds, ester compounds, sulfate compounds, and sulfite compounds; and the term "lithium protection solvent", as used herein, is defined as a solvent that forms a good protective layer, i.e. a stable solid-electrolyte interface (SEI) layer, on a lithium surface, and which shows a cyclic efficiency of at least 50%, and the lithium protection solvent is selected from saturated ether compounds, unsaturated ether compounds, and heterocyclic compounds including N, O, and/or S.

Examples of the weak polar solvents include xylene, dimethoxyethane, 2-methyltetrahydrofuran, diethyl carbonate, dimethyl carbonate, toluene, dimethyl ether, diethyl ether, diglym, and tetraglyme.

Examples of the strong polar solvents include hexamethyl phosphoric triamide, γ - butyrolactone, acetonitrile, ethylene carbonate, propylene carbonate, N-methyl pyrrolidone, 3-methyl-2-oxazolidone, dimethyl formamide, sulfolane, dimethyl acetamide, dimethyl sulfate, ethylene glycol diacetate, dimethyl sulfite, and ethylene glycol sulfite.

Examples of the lithium protection solvents include tetrahydrofuran, 1,3-dioxolane, 3,5-dimethylisoxazole, 2,5-dimethyl furan, furan, 2-methyl furan, 1,4-oxane, and 4-methyldioxolane.

Examples of electrolyte salts include lithium trifluoromethane sulfonimide, lithium triflate, lithium perchlorate, LiPF₆, LiBF₄, tetraalkylammonium salts, such as tetrabutylammonium tetrafluoroborate (TBABF₄), liquid state salts at room temperature, *e.g.* imidazolium salts such as 1-ethyl-3-methylimidazolium Bis-(perfluoroethyl sulfonyl) imide (EMIBeti), and combinations thereof.

The following examples illustrate the present invention in further detail, but it is understood that the present invention is not limited by these examples.

Example 1

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 V_2O_5 powder was pulverized in an isopropyl alcohol solvent with a zirconia ball for 6 hours, and the resulting material was dried at 80°C to prepare a V_2O_5 additive with a particle size D (v, 50%) of 200 nm.

The V_2O_5 additive, an elemental sulfur (S_8) positive active material, a carbon conductive material, and a polyethyleneoxide binder were mixed in an acetonitrile solvent in the weight ratio of 1:6:2:2 with a ball to prepare a positive active material slurry. The elemental sulfur (S_8) was obtained from pulverization with a zirconia ball in an isopropylalcohol solvent and drying, and it had a particle size D (v, 50%) of 5,000 nm.

The positive active material slurry was coated on a carbon-coated Al current collector to produce a positive electrode for a lithium-sulfur battery.

Example 2

A positive electrode was produced by the same procedure as in Example 1, except that the pulverization step was performed for 12 hours to prepare a V_2O_5 additive with a particle size D (v, 50%) of 50 nm.

Example 3

A positive electrode was produced by the same procedure as in Example 1, except that the pulverization step was performed for 24 hours to prepare a V_2O_5 additive with a particle size D (v, 50%) of 10 nm.

Comparative Example 1

A positive electrode was produced by the same procedure as in Example 1, except that the pulverization step was performed for 1 hour to prepare a V_2O_5 additive with a particle size D (v, 50%) of 3,000 nm.

Comparative Example 2

A positive electrode was produced by the same procedure as in Example 1, except that the V_2O_5 additive with a particle size D (v, 50%) of 150,000 nm without a pulverization step was used.

Comparative Example 3

A positive electrode was produced by the same procedure as in Example 1, except that a V_2O_5 additive was not used.

Experiment 1: Measurement of ionic conductivity according to a particle size D (v, 50%) of V₂O₅>

Polyethylene oxide with a molecular weight of 5,000,000 was dissolved in acetonitrile, and a $LiN(SO_2CF_3)_2$ lithium salt was added thereto and dissolved therein until the mole ratio of ethylene oxide to Li reached 10 : 1. The V_2O_5 additive according to Examples 1 to 3 and Comparative Examples 1 to 2 was added to the resulting solution in an amount of 10 wt% based on the total amount of polyethylene oxide and the $LiN(SO_2CF_3)_2$ lithium salt, and they were shaken for 2 hours. The resulting solution was cast to form a polymer film, and the ionic conductivity thereof was measured. The results are presented in Table 1.

Table 1

Type of polymer film	Ionic conductivity (S/cm) (room temperature)	
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂	9.6 × 10 ⁻⁷	
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (D(v, 50%)=150,000nm)	1.0 × 10 ⁻⁶	
PEO_{10} -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (D(v, 50%)=30,000nm)	2.5 × 10 ⁻⁶	
PEO_{10} -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (D(v, 50%)=200nm)	8.6 × 10 ⁻⁶	
PEO_{10} -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (D(v, 50%)=50nm)	3.0 × 10⁻⁵	
PEO_{10} -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (D(v, 50%)=10nm)	5.0 × 10 ⁻⁵	

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As shown in Table 1, the addition of V_2O_5 increased ionic conductivity, and such an increase in ionic conductivity is improved as the particle size of V_2O_5 decreases. This believed to be because the inorganic additive such as V_2O_5 prevents crystallization of the polymer.

Increases in ionic conductivity according to the amount of V_2O_5 were measured. The V_2O_5 with a particle size D (v, 50%) of 10 nm was used. The results are presented in Table 2.

Table 2

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Type of polymer film	lonic conductivity (S/cm)(room temperature)
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (2 wt%)	(S/cm)(room temperature) 1.0 × 10 ⁻⁵
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (5 wt%)	4.0 × 10 ⁻⁵
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (10 wt%)	5.0 × 10 ⁻⁵
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (15 wt%)	3.5 × 10 ⁻⁵
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (20 wt%)	2.0 × 10 ⁻⁵
PEO ₁₀ -LiN(SO ₂ CF ₃) ₂ -V ₂ O ₅ (25 wt%)	8.0 × 10 ⁻⁶

It is evident from Table 2 that the amount of 5 to 15 wt% of V_2O_5 exhibited higher ionic conductivity.

These two test results indicate that V_2O_5 increases ionic conductivity, and as the particle size of V_2O_5 decreases, ionic conductivity increases.

< Experiment 2: Measurement of surface roughness>

The surface roughness Ra of the positive electrodes according to Examples 1 to 3 and Comparative Examples 1 to 3 were measured, and the results are shown in Table 3.

Table 3

	Composition of the positive electrode (weight ratio)	Surface roughness (Ra)
Comparative Example 3	Sulfur/Conductive material/Binder (6/2/2)	3.0
Comparative Example 1	Sulfur/Conductive material/Binder/ V_2O_5 (A: D(v, 50%)=150,000nm) (6/2/2/1)	12.6
Comparative Example 2	Sulfur/Conductive material/Binder/V ₂ O ₅ (A: D(v, 50%)=30,000nm) (6/2/2/1)	6.6
Example 1	Sulfur/Conductive material/Binder/V ₂ O ₅ (A: D(v, 50%)=200nm) (6/2/2/1)	2.9
Example 2	Sulfur/Conductive material/Binder /V ₂ O ₅ (A: D(v, 50%)=50nm) (6/2/2/1)	2.8
Example 3	Sulfur/Conductive material/Binder /V ₂ O ₅ (A: D(v, 50%)=10nm) (6/2/2/1)	2.5

In Table 3, Ra indicates the arithmetic mean of each peak (between highest and lowest peaks), and the lower Ra indicates a more uniform surface. It is evident from Table 3 that Comparative Examples 1 and 2 with the particle size of V_2O_5 larger than the sulfur active material (5,000 nm) exhibited poorer uniformity (unevenness and roughness) than Comparative Example 3 without V_2O_5 , and Examples 1 to 3 with a smaller or the same particle size of V_2O_5 as the active material exhibited substantially the same or better uniformity.

< Experiment 4: Measurement of battery performance>

Using positive electrodes according to each of Examples 1 to 3 and Comparative Examples 1 to 3, pouch-type lithium-sulfur cells were fabricated by the following procedure. The size of each positive electrode was 25mm X 50mm. The cells were test cells with a higher capacity than a coin cell (capacity of 3-5mAh).

A tab was welded to each positive electrode and the resulting positive electrode was placed in a pouch. On the positive electrode, a separator was positioned. A tab-attached lithium foil was placed on the separator, and the pouch was sealed except for at an electrolyte inserting hole. 1M LiN(SO₂CF₃)₂ in dimethoxyethane/1,3-dioxolane (80/20 volume ratio) was injected into the pouch.

The cell was charged at 0.2C and discharged at 0.5C, the 1st capacity and cycle life for 100th cycles were measured, and the results are presented in Table 4. In addition, internal resistance of the battery and surface roughness Ra of the positive electrode are shown in Table 4.

Table 4

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	Surface roughness (Ra)	Internal resistance (Ω)	1 st capacity (mAh/g)	Cycle life for 100th cycles (%)
Comparative Example 3	3.0	9.8	1200	60
Comparative Example 1	12.6	15.3	1053	55
Comparative Example 2	6.6	12.6	1125	58
Example 1	2.9	9.5	1230	85
Example 2	2.8	9.6	1250	88
Example 3	2.5	9.5	1280	90

It is shown in Table 4 that Examples 1 to 3 using V_2O_5 with the same or smaller particle size than the sulfur positive active material (5,000 nm) exhibited lower surface roughness than Comparative Example 3 without V_2O_5 . Such a lower surface roughness renders a decrease in

interfacial resistance, causing a decrease in internal resistance of the battery and an increase in 1st capacity and cycle life.

On the other hand, Comparative Examples 1 and 2 using V_2O_5 with a particle size larger than the sulfur positive active material (5,000 nm) exhibited greater surface roughness, which results in an increase in internal resistance and a decrease in capacity and cycle life.

Examples 4 to 8: Test for battery performance according to amount of added V₂O₅

Positive electrodes were produced by the same procedure as in Example 1, except that amounts of V_2O_5 with a particle size D (v, 50%) of 10 nm were changed as set forth in the following Table 5. The surface roughness of each positive electrode was measured, and the results are shown in Table 5. The result according to Example 4 with a particle size D (v, 50%) of 10 nm is also shown in Table 5.

Table 5

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	Composition of positive electrode (weight ratio)	Surface roughness (Ra)
Comparative Example 3	Sulfur/Conductive material/Binder (60/20/20)	3.0
Example 4	Sulfur/Conductive material/Binder/V₂O₅(A: D(v, 50%)=10nm) (60/20/20/2)	2.9
Example 5	Sulfur/Conductive material/Binder/V₂O₅(A: D(v, 50%)=10nm) (60/20/20/5)	2.8
Example 3	Sulfur/Conductive material/Binder/V₂O₅(A: D(v, 50%)=10nm) (60/20/20/10)	2.5
Example 6	Sulfur/Conductive material/Binder/V₂O₅(A: D(v, 50%)=10nm) (60/20/20/15)	2.4
Example 7	Sulfur/Conductive material/Binder/V ₂ O ₅ (A: D(v, 50%)=10nm) (60/20/20)	2.6
Example 8	Sulfur/Conductive material/Binder/V ₂ O ₅ (A: D(v, 50%)=10nm) (60/20/25)	2.5

Table 4 indicates that the same or smaller particle size of V_2O_5 according to Example 3 to 8 than the positive active material decreases surface roughness when compared with Comparative Example 1 without V_2O_5 .

< Experiment 5: Measurement of Battery Performance>

Using positive electrodes according to each of Examples 3 to 8, pouch-type lithium sulfur cells were fabricated by the same procedure as in Experiment 4. The cells were charged at 0.2C and discharged at 0.5C, the 1st capacity and cycle life for the 100th cycles were measured, and the results are presented in Table 6. In addition, internal resistance of each battery and surface roughness Ra of each positive electrode are shown in Table 6.

Table 6

	Surface roughness (Ra)	Internal resistance (Ω)	1 st capacity (mAh/g)	Cycle life for 100 th cycles (%)
Comparative Example 3	3.0	9.8	1200	60
Example 4	2.9	9.7	1220	70
Example 5	2.8	9.5	1245	88
Example 3	2.5	9.5	1280	90
Example 6	2.4	9.3	1254	84
Example 7	2.6	9.4	1234	82
Example 8	2.5	9.3	1230	7 2

It is shown in Table 6 that Examples 3 to 8 using V_2O_5 with the same or smaller particle size than the sulfur positive active material (5,000 nm) exhibited lower surface roughness than Comparative Example 1 without V_2O_5 . Such a lower surface roughness renders a decrease in interfacial resistance, causing a decrease in internal resistance of the battery. As a result, the 1st capacities in Examples are slightly larger than that in Comparative Example 3, and the cycle life greatly increased by 5 to 20%. This is believed to result from higher ionic conductivity in 5 to 20wt% of V_2O_5 , even though Comparative Example have similar surface roughness (See Table 2).

[Effect of the invention]

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As a result, the V_2O_5 additive with the critical particle size increases ionic conductivity of the positive electrode and decreases surface roughness, thereby increasing capacity from 1200mAh to 1280mAh, and improving cycle life from 60% to 90%.

WHAT IS CLAIMED IS:

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- 1. A positive electrode for a lithium-sulfur battery comprising:
- a positive active material selected from the group consisting of elemental sulfur (S_8) , a sulfur-based compound, and mixtures thereof;
 - a conductive material;
 - a binder; and
 - a V_2O_5 additive with a particle size D (v, 50%) of 1,000nm or less.
- 2. The positive electrode of claim 1, wherein the inorganic additive has a particle size D (v, 50%) of 1 to 900 nm.
- 3. The positive electrode of claim 2, wherein the inorganic additive has a particle size D (v, 50%) of 5 to 700 nm.
- 4. The positive electrode of claim 3, wherein the inorganic additive has a particle size D (v, 50%) of 10 to 500 nm.
- 5. The positive electrode of claim 1, wherein a surface roughness of the positive electrode, Ra, is $5 \mu m$ or less.
- 6. The positive electrode of claim 5, wherein a surface roughness of the positive electrode, Ra, is 0.1 to $5 \mu m$.
- 7. The positive electrode of claim 6, wherein a surface roughness of the positive electrode, Ra, is 0.2 to 4 μm .
- 8. The positive electrode of claim 1, wherein the V_2O_5 additive is present in an amount of 1 to 50 wt%.
- 9. The positive electrode of claim 1, wherein the V_2O_5 additive is present in an amount of 2 to 25 wt%.
- 10. The positive electrode of claim 1, wherein the V_2O_5 additive is present in an amount of 3 to 20 wt%.
- 11. The positive electrode of claim 1, wherein the sulfur-based compound is selected from the group consisting of Li_2S_n , wherein $n \ge 1$,) organic-sulfur compounds, and carbon-sulfur polymers having the formula $(C_2S_x)_n$, where x=2.5 to 50 and $n \ge 2$.
- 12. The positive electrode of claim 1, wherein the positive electrode further comprises a coating layer, the coating layer comprising a polymer, an inorganic material, or a mixture thereof.

- 13. The positive electrode of claim 12, wherein the coating layer comprises a polymer selected from the group consisting of polyvinylidene fluoride, copolymers of polyvinylidene fluoride and hexafluoropropylene, poly(vinyl acetate), poly(vinyl butyral-co-vinyl alcohol-co-vinyl acetate), poly(methylmethacrylate-co-ethyl acrylate), polyacrylonitrile, polyvinyl chloride-co-vinyl acetate, polyvinyl alcohol, poly(1-vinylpyrrolidone-co-vinyl acetate), cellulose acetate, polyvinyl pyrrolidone, polyacrylate, polymethacrylate, polyolefin, polyurethane, polyvinyl ether, acrylonitrile-butadiene rubber, styrene-butadiene rubber, acrylonitrile-butadiene styrene, a sulfonated styrene/ethylene-butylene/styrene triblock copolymer, polyethylene oxide, and mixtures thereof.
- 14. The positive electrode of claim 12, wherein the coating layer comprises an inorganic material selected from the group consisting of colloidal silica, amorphous silica, surface-treated silica, colloidal alumina, amorphous alumina, tin oxide, titanium oxide, vanadium oxide, titanium oxide (TiS₂), zirconium oxide (ZrO₂), iron oxide, iron sulfide (FeS), iron titanate (FeTiO₃), barium titanate (BaTiO₃), and mixtures thereof.
 - 15. A lithium sulfur-battery comprising:

a positive electrode comprising a positive active material selected from the group consisting of elemental sulfur (S_8), sulfur-based compounds, and mixtures thereof, a conductive material, a binder and an V_2O_5 additive with a particle size D (v, 50%) of 1,000nm or less;

a negative electrode comprising a negative active material selected from the group consisting of a material that is capable of reversibly intercalating or deintercalating lithium ions, a material that reacts with lithium ions to prepare a lithium-included compound, a lithium metal, and a lithium alloy; and

an electrolyte.

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- 16. The lithium-sulfur battery of claim 15, wherein the V_2O_5 additive has a particle size D (v, 50%) of 1 to 900 nm.
- 17. The lithium-sulfur battery of claim 16, wherein the V_2O_5 additive has a particle size D (v, 50%) of 5 to 700 nm.
- 18. The lithium-sulfur battery of claim 16, wherein the inorganic additive has a particle size D (v, 50%) of 10 to 500 nm.
- 19. The lithium-sulfur battery of claim 15, wherein a surface roughness of the positive electrode, Ra, is 5 µm or less.
- 20. The lithium-sulfur battery of claim 19, wherein a surface roughness of the positive electrode, Ra, is 0.1 to $5 \, \mu m$.

- 21. The lithium-sulfur battery of claim 20, wherein a surface roughness of the positive electrode, Ra, is 0.2 to $4 \mu m$.
- 22. The lithium-sulfur battery of claim 15, wherein the positive electrode further comprises a coating layer, the coating layer comprising a polymer, an inorganic material or a mixture thereof.

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- 23. The lithium-sulfur battery of claim 22, wherein the coating layer comprises a polymer selected from the group consisting of polyvinylidene fluoride, copolymers of polyvinylidene fluoride and hexafluoropropylene, poly(vinyl acetate), poly(vinyl butyral-co-vinyl alcohol-co-vinyl acetate), poly(methylmethacrylate-co-ethyl acrylate), polyacrylonitrile, polyvinyl chloride-co-vinyl acetate, polyvinyl alcohol, poly(1-vinylpyrrolidone-co-vinyl acetate), cellulose acetate, polyvinyl pyrrolidone, polyacrylate, polymethacrylate, polyolefin, polyurethane, polyvinyl ether, acrylonitrile-butadiene rubber, styrene-butadiene rubber, acrylonitrile-butadiene styrene, a sulfonated styrene/ethylene-butylene/styrene triblock copolymer, polyethylene oxide, and mixtures thereof.
- 24. The lithium-sulfur battery of claim 22, wherein the coating layer comprises an inorganic material selected from the group consisting of colloidal silica, amorphous silica, surface-treated silica, colloidal alumina, amorphous alumina, tin oxide, titanium oxide, vanadium oxide, titanium oxide (TiS₂), zirconium oxide (ZrO₂), iron oxide, iron sulfide (FeS), iron titanate (FeTiO₃), barium titanate (BaTiO₃), and mixtures thereof.